UV and Visible Harmonic Generation Through Enhanced Transmission from GaAs-filled Nanocavities

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Abstract

A theoretical study of harmonic generation from a silver grating having slits filled with GaAs has been conducted. The enhanced transmission regime that guarantees high field localization inside the slits, and the phase-locking mechanism that take place between the pump and its harmonics allows enhanced harmonic generation under conditions of high absorption at visible and UV wavelengths.

1. Introduction

Since the first observation of enhanced optical transmission (EOT) [1], efforts have multiplied to prove that this condition coincides with strong field localization on the metal surface and in proximity of the apertures [2], suggesting also that these structures can be a good mean to achieve enhanced nonlinear phenomena. However, since metals are centrosymmetric and lack a second order nonlinear term their nonlinear response comes mostly from the third order nonlinear term and from a combination of symmetry breaking at the surface and from volume contributions [3, 4]. The ability of slits to support TEM-like resonant modes [5] allows more opportunities to efficiently generate harmonic fields when the apertures are filled with nonlinear materials [6]. However, a detailed analysis of dynamical contributions of the metal to SHG and THG from electron gas pressure, convection, inner core electrons and a $\chi^{(3)}$ response and phase-locking between the pump and the harmonics that allows generation in wavelength ranges below the absorption edge has never been conducted. This study shows harmonic generation in visible and UV ranges for metal grating filled with nonlinear materials without imposing any separation between surface and volume sources [7, 8]. We described conduction electrons in metal by including Coulomb (electric), Lorentz (magnetic), convective, electron gas pressure and linear and nonlinear contributions to the dielectric constant of the metal arising from bound electrons [9].

2. Linear and Nonlinear Results

We begin our analysis by examining the behavior of a single slit of size *a* filled with GaAs, and carved on a silver layer having thickness *w*. The FF is tuned in a transparency region (ε_{GaAs}(1064nm) ~ 12.10), while both second (532nm) and third harmonic (354nm) are tuned deep in the absorbing region (respectively ε_{GaAs}(532nm)~17.08+ i2.86 and ε_{GaAs}(354nm)~8.81+ i14.36), where no harmonic generation is expected. In order to favor nonlinear processes we optimized the linear transmission properties using incident TM-polarized light: by varying thickness and aperture size we obtained a transmission map that reveals the strong resonant nature of the structure (Fig. 1(a)). Further enhancement of the linear response can be achieved by arranging the slit in a periodic pattern. The simulations carried out on an infinite array of slits 60nm wide on a 100nm-thick silver film, where the periodicity has been varied from p = 200nm to p = 3200nm (Fig.1(b)), shows that the role of array periodicity (or pitch size) can be detrimental if its value is a multiple of the surface plasmon wave-

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14. ABSTRACT
A theoretical study of harmonic generation from a silver grating having slits filled with GaAs has been conducted. The enhanced transmission regime that guarantees high field localization inside the slits, and the phase-locking mechanism that take place between the pump and its harmonics allows enhanced harmonic generation under conditions of high absorption at visible and UV wavelengths.

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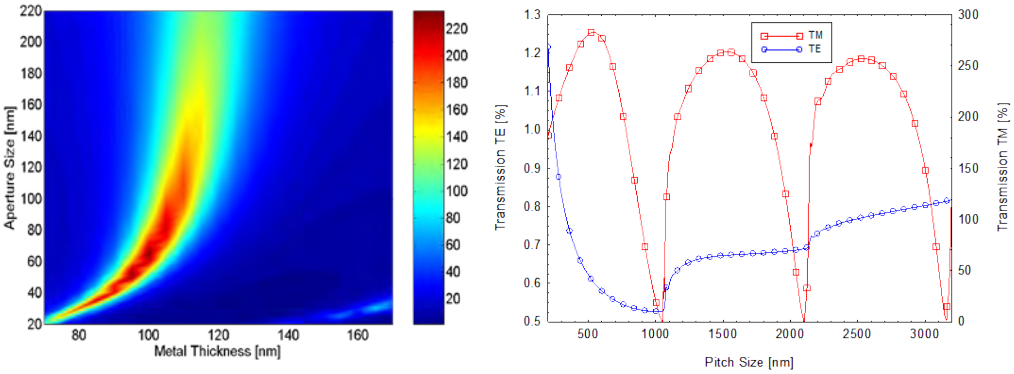
length of the dielectric/metal unperturbed interface [10]. The interference of horizontal resonances with the modes resonating in the vertical direction favors strong modulation of the linear transmission profile (Fig.1(b)), causing the appearance of a gap every time the surface plasmon wavelength matches the periodicity of the array. Transmission values for an incident, TE-polarized pump field – blue line in Fig.1(b) – are less than 1% for large periodicities, and approach 1% when slit-to-slit-distance is relatively small. The reason for the enormous difference between the two polarizations is related to the lack of resonant Fabry-Perot modes for TE-polarized light, which at 1064nm is well below the cut-off. Moreover TE-polarized light also exhibits strong transmission minima due to the interference of horizontal and vertical resonances. However, while vertical modes for TM-polarized light can be ascribed to the coupling and back-radiation of surface plasmons on the impinging interface, these modes change their nature for a TE-polarized field, matching exactly the Rayleigh minimum condition.

Since the enhanced transmission process is always characterized by field localization, absorption and field penetration inside the metal (in these ranges transition metals display dielectric constants of order unity), the interaction of light with both free and bound electrons in metals becomes more efficient especially if the light is concentrated and enhanced in small volumes. Moreover, when a material having non negligible $\chi^{(2)}$ and/or $\chi^{(3)}$ values fills the slits, new channels for harmonic generation become available and eventually lead to phase-locked pump photon down conversion. We illuminated the array described above with pulses approximately 120fs in duration, with peak intensities of roughly 2GW/cm^2 and calculated the nonlinear response considering bound and free electrons contribution arising from the metal, and bound electrons from GaAs, modeled as outlined in Ref. 26. We considered quadratic and cubic nonlinear terms for GaAs only. An impinging TM-polarized field generates four nonlinear cross-polarized harmonic fields: TM- and TE-polarized SH and TH. The generated fields reveal the dramatic influence of the linear response on the nonlinear one for both polarizations: all the generated harmonics experience the same forbidden states as the incident pump field does. Moreover, the phase locking process [11-12] is playing a non trivial role in harmonic generation. A 100nm-thick GaAs substrate is only 20% transparent at 532nm, and completely opaque at 354nm. More convincing numerical evidence of phase locking may be achieved by increasing substrate thickness to ~170nm, and by reducing the width of the nano-channel down to 20nm, so that we are still operating under resonant conditions. The result is that conversion efficiencies do not vary significantly, even though all the TE-generated harmonics are now far below cut-off. This is a sure sign that phase locking is the main mechanisms that drives the harmonic field to resonate inside the cavity even if it is tuned to resonate at the pump frequency. It is worth noting that the down-conversion to TE-polarized pump photons (see Fig.2) is not trivial because the transmission of an incident TE-polarized pump field in this structure should be completely forbidden, as waveguide theory suggests and Fig.1(b) demonstrates.



(a) (b)

Fig.1. (a) Transmission map at λ=1064nm for a single slit carved on a silver substrate, filled with a material having ε_{GaAs}=12.10+ i0. (b) Transmission versus pitch size at 1064nm for both TM (red line – square markers, right axis) and TE (blue line – circle markers, left axis) polarization.

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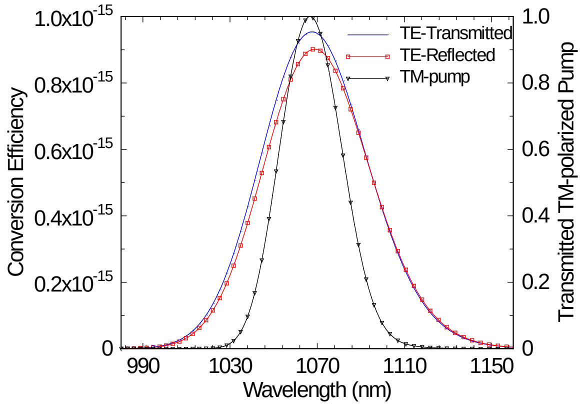

Fig.2: TE-polarized down-converted pump photon efficiency transmitted (blue line – circle markers) and reflected (red line – square markers) from an array of slits 60nm wide, filled with GaAs. The spectrum of the TE pump is compared with the incident TM pump (black line – triangle markers). Silver thickness is 100nm and array periodicity has been fixed to p= 590nm.

4. Conclusion

Second and third harmonic generation, as well as cross-polarized down conversion processes from GaAs filled sub-wavelength slits have been demonstrated using a general model that allows to analyze linear and nonlinear dynamics without making any assumptions about either the roles or quantitative contribution of each type of nonlinear source. Harmonic generation in both polarizations has been shown to be possible thanks to the phase locking mechanism that takes place even in the enhanced transmission regime.

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